Nonlinear Terahertz Responses of Single-Wall Carbon Nanotube Films

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Single-wall carbon nanotubes (SWCNT) have a versatile, chirality-dependent electronic character and unique one-dimensional (1D) properties that are ideal for optoelectronic applications. Metallic SWCNTs have a gapless and massless electronic dispersion akin to parent material graphene while semiconducting SWCNTs have diameter-dependent direct band gaps. Although their linear optical properties have been extensively studied over the past decade, their nonlinear optical properties remain largely unexplored, particularly in the terahertz (THz) range. Here, we investigate the nonlinear THz response of aligned and unaligned SWCNT films of various chirality compositions. Specifically, we focused on third harmonic generation (THG) arising from the third-order conductivity of 1D massless carriers. We first utilized Fourier-transform infrared spectroscopy and time-domain THz spectroscopy (TDTS) to characterize the film's linear low-energy dynamics and provide initial insights. For nonlinear spectroscopy, we employed an alternate TDTS setup producing large THz electric field pulses using the tilted-pulse-front technique. Drawing on recently published predictions of third-order quantum behavior in the intraband dynamics of massless carriers in graphene, we expect strongly anisotropic nonlinear responses of 1D carriers in metallic SWCNT films due to their similar, massless band structure. Conversely, we do not expect to observe THG in semiconducting SWCNTs, which lack free carriers. This study presents a unique contribution to the field by offering new insights in fundamental 1D solid-state physics while also giving rise to a new class of efficient, high-frequency THz emitters based on prototypical low-dimensional materials systems.

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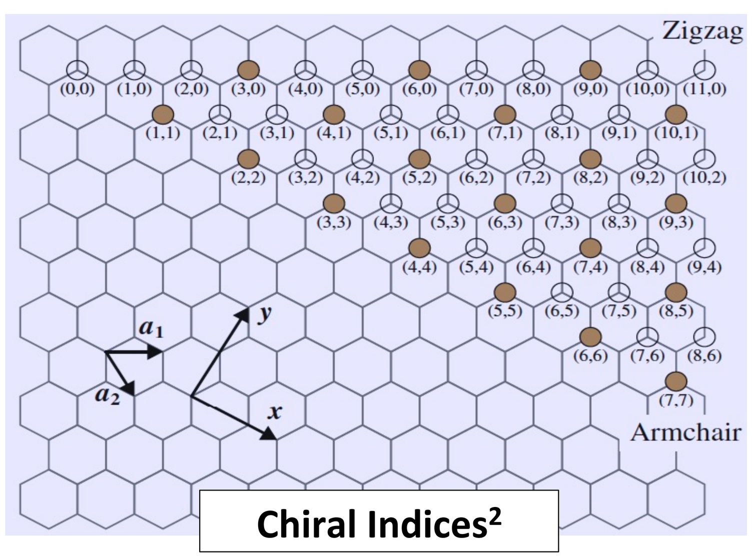
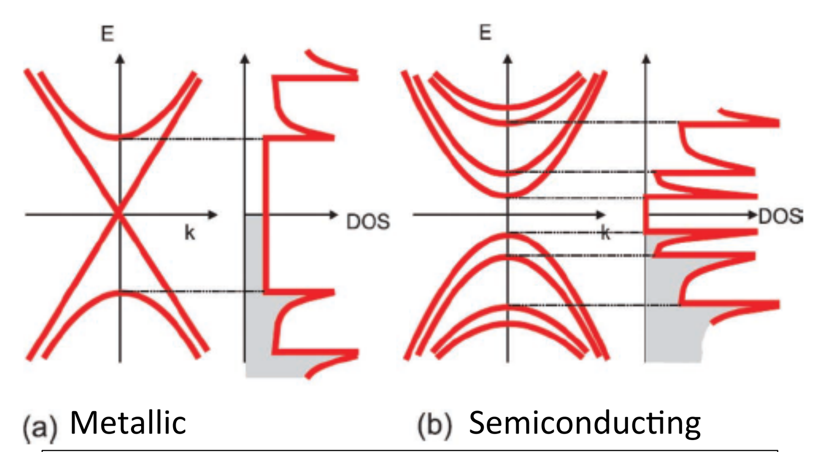
Introduction

Single-wall carbon nanotubes (SWCNT): Rolled-up graphene (*sp^2-hybridized C*)

- Unique one-dimensional (1D) properties ideal for optoelectronic applications[1]
- Chirality-dependent electronic character: $C_h = na_1 + ma_2 \rightarrow$ chiral index (n,m)
  For [(n – m) mod 3 = 0: metallic, otherwise semiconducting SWCNT



Chiral Indices[2]



Electronic Dispersion and Density of States (DOS) of SWCNT[1]

- Metallic SWCNT have massless, gapless electronic dispersion like graphene
- Nonlinear optical response is unexplored in 1D SWCNT particularly in the terahertz (THz) range

Goal: Investigate the nonlinear THz response of SWCNT films to observe third harmonic generation (THG), induced transparency or absorption saturation

Methods

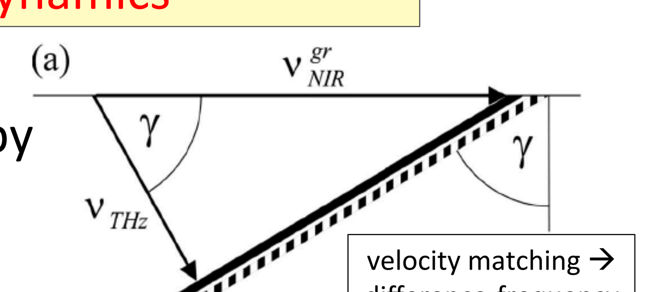
THz and infrared spectroscopy are powerful tools for probing low-energy carrier dynamics

Characterizing the linear THz/MIR response:

- Fourier-transform infrared (FTIR) spectroscopy
- Time-domain terahertz spectroscopy (TDTS)

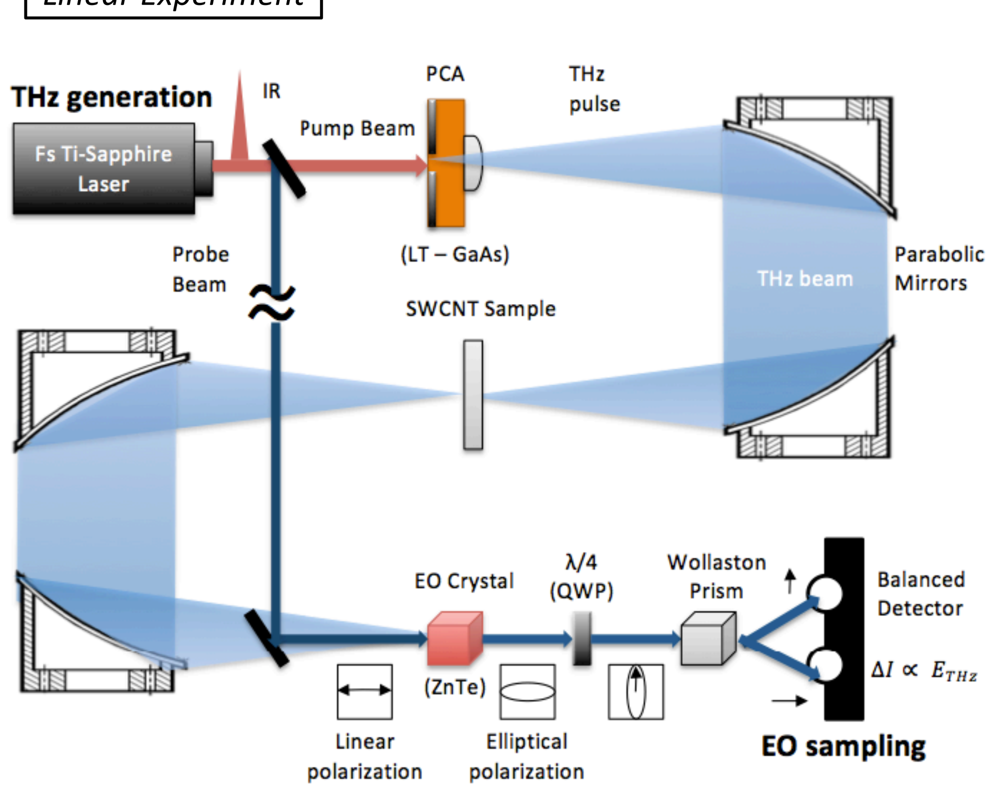
Characterizing the nonlinear THz response:

- TDTS with powerful tilted-pulse-front technique[3]

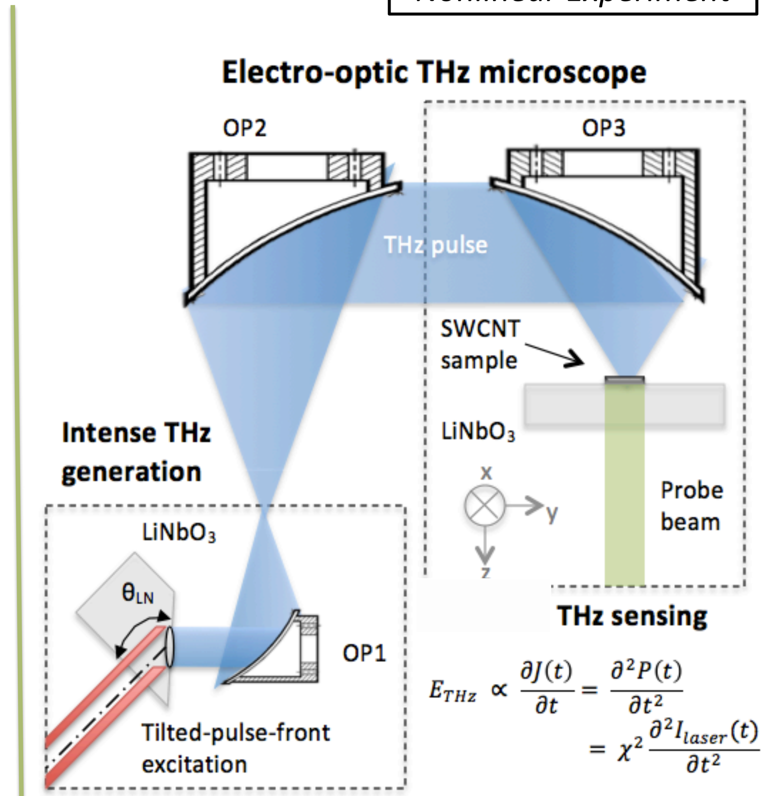


TDTS Experimental Setup

Linear Experiment



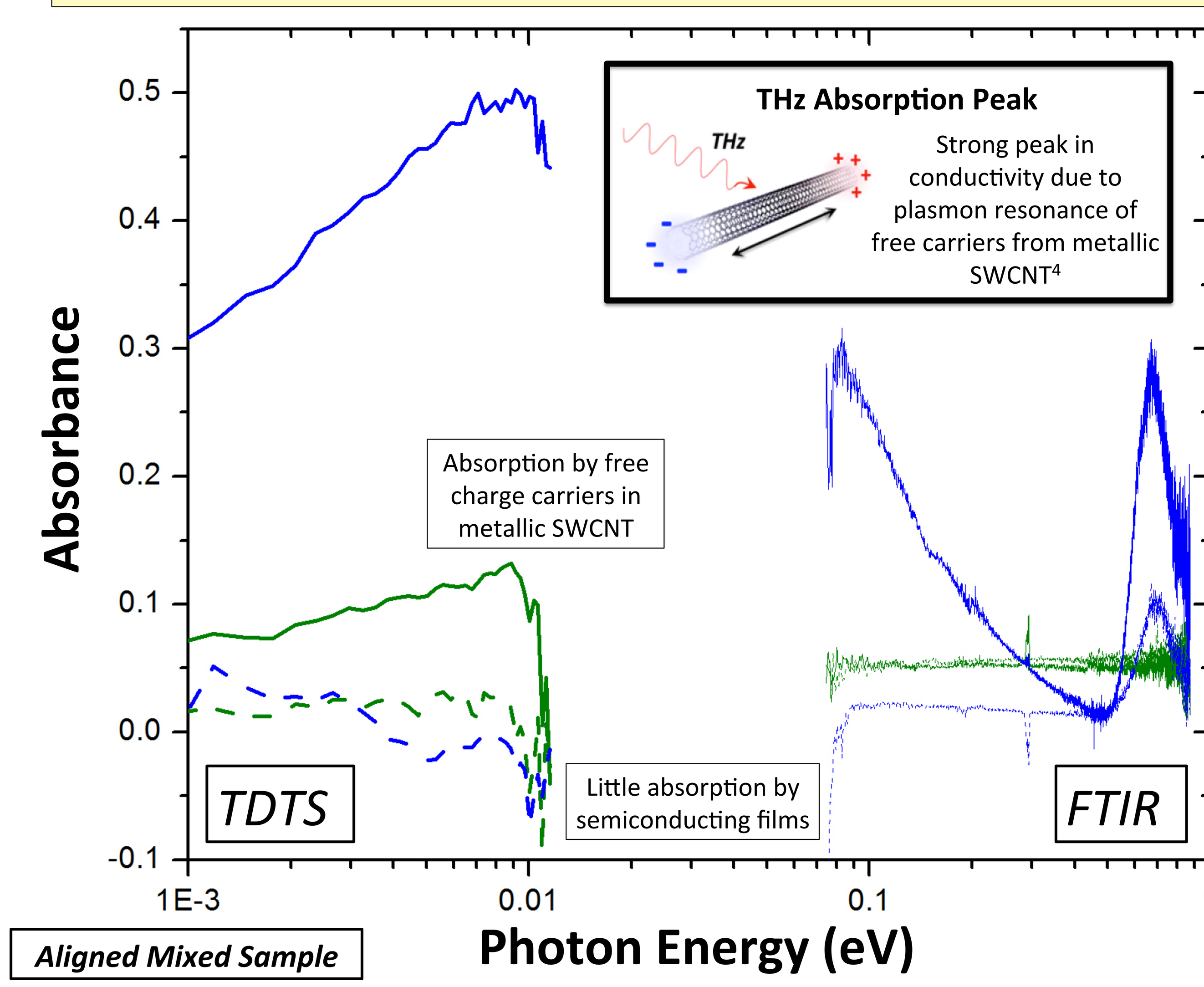
Nonlinear Experiment



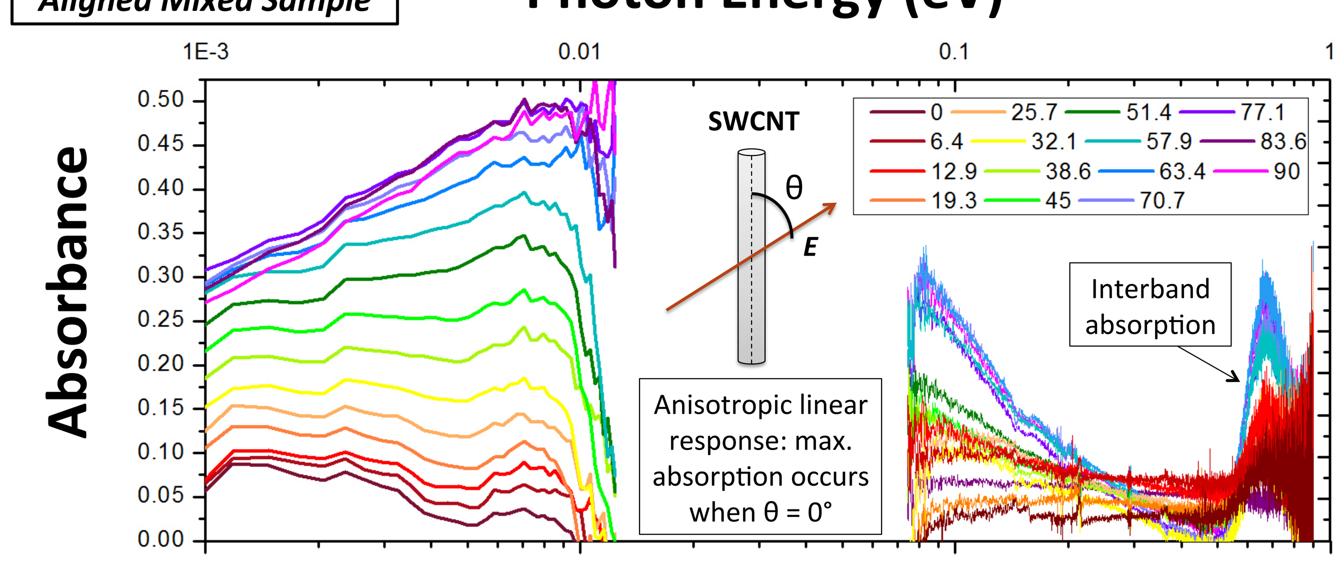
Results

Linear THz Response

4 samples of different alignment and chirality composition:
Unaligned (6,6) — Aligned Mixed
Unaligned (6,5) — Aligned Semiconducting

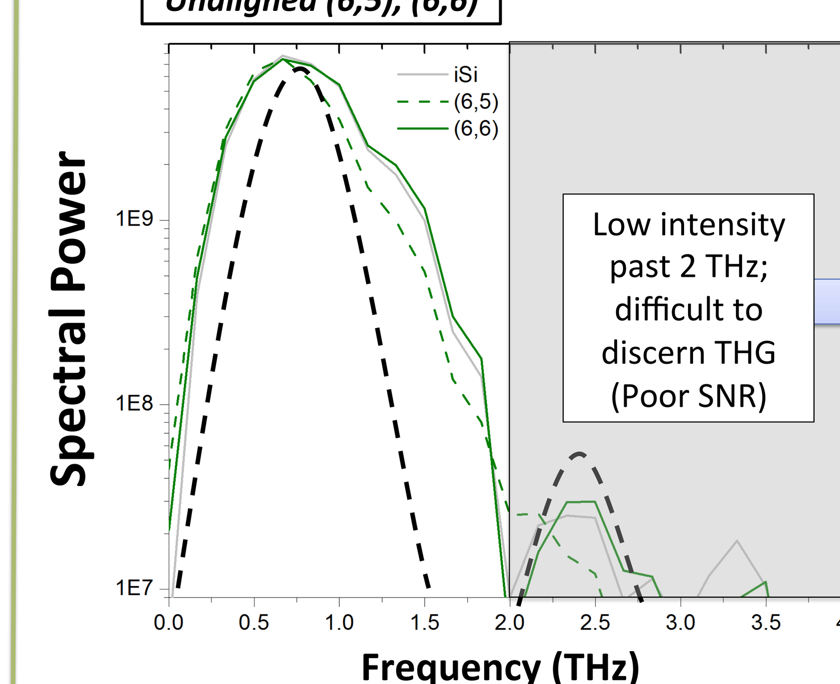


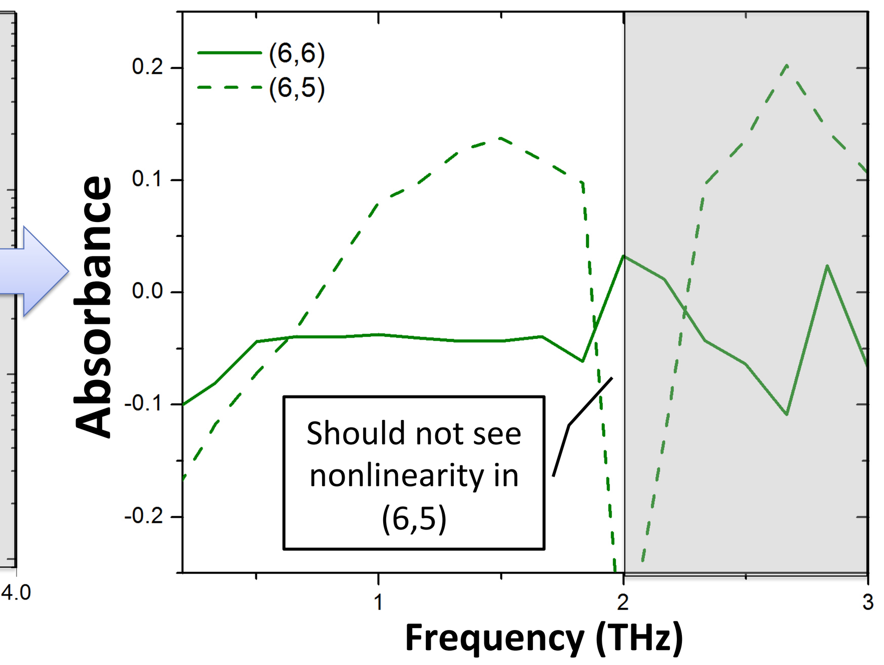
Aligned Mixed Sample



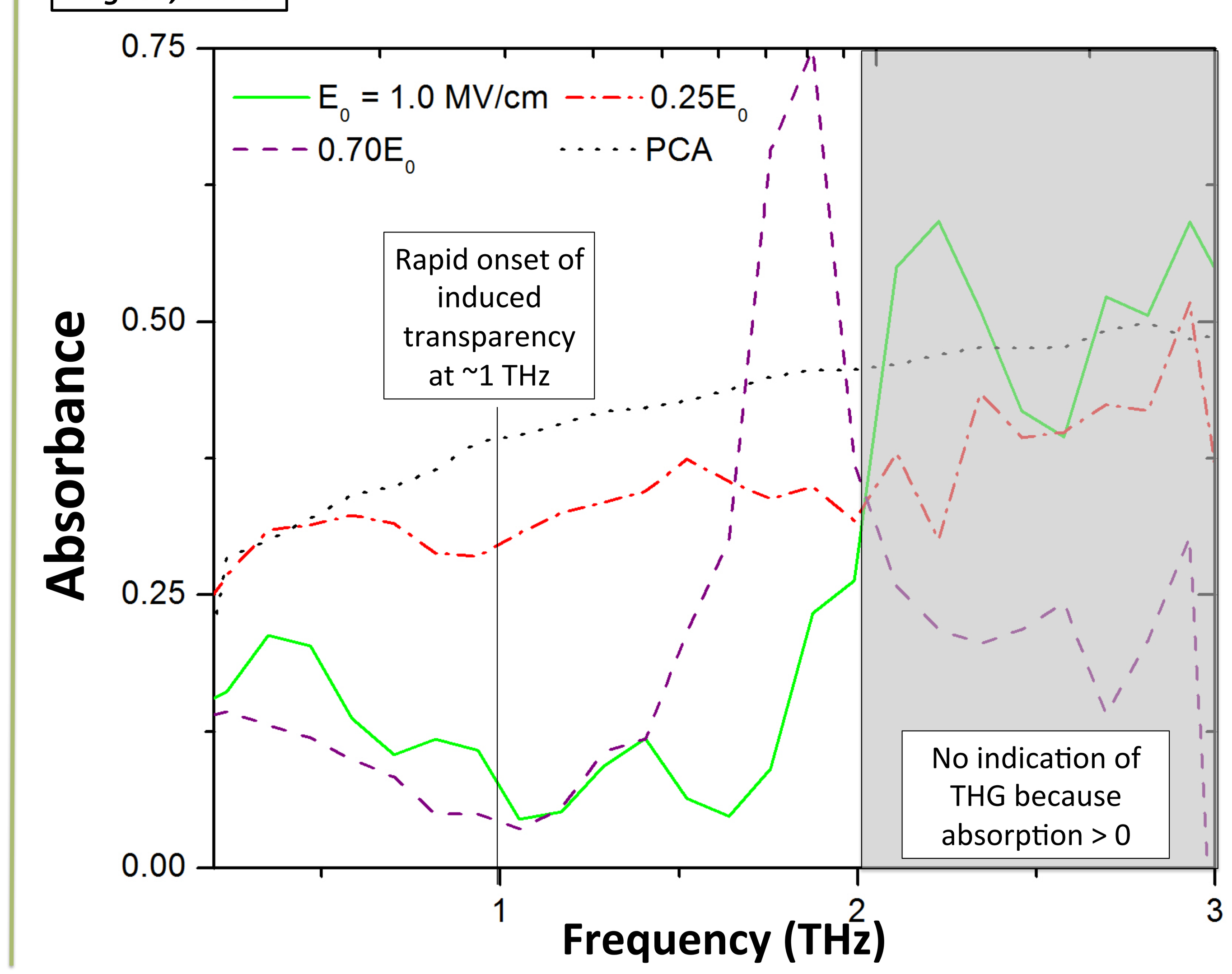
Nonlinear THz Response

Unaligned (6,5), (6,6)

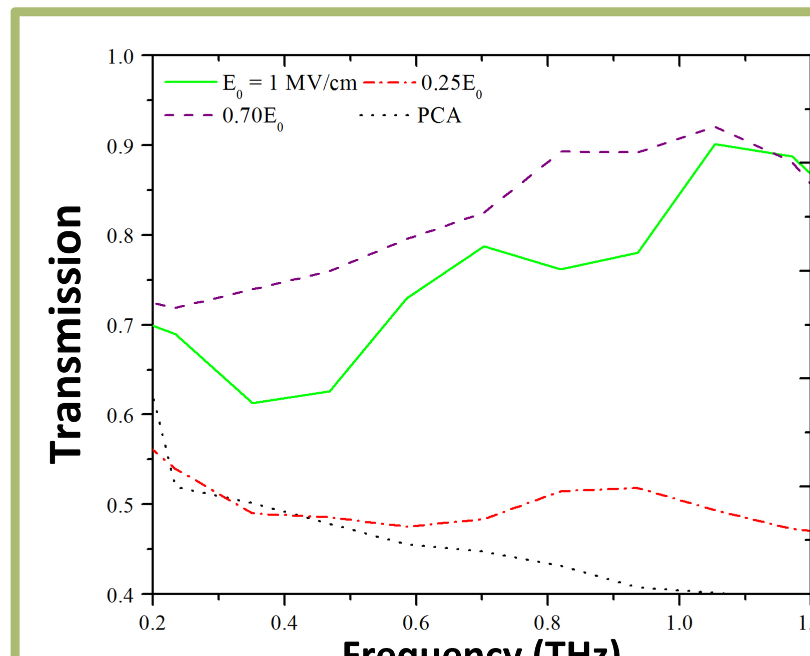




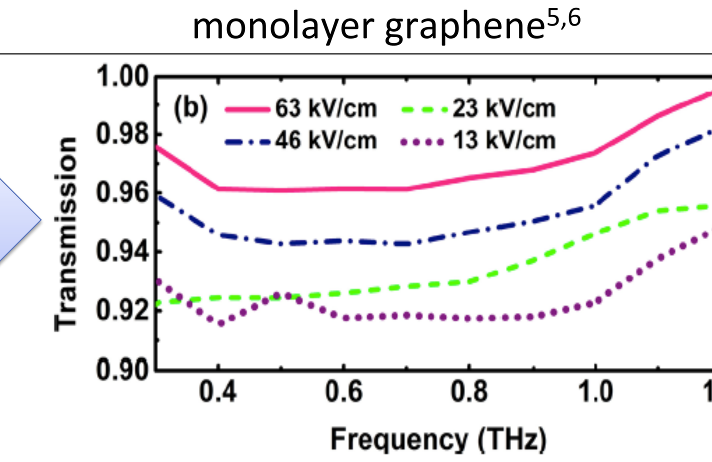
Aligned, Mixed



Discussion



Maximum transmission increase of 42.7% from 0.25 to 1.0 MV/cm at ~1 THz with "threshold behavior" observed in monolayer graphene[5,6]



Conclusions

- Strong nonlinear behavior visible in SWCNT with free carriers at ~ 1 THz
- Increased transmission at high electric field strength bears phenomenological resemblance to recent studies of graphene
- Further investigation of induced transparency in enriched SWCNT is necessary

Acknowledgements

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